

522 Rec'd PCT/PTD 01 MAR 2000

Form PTO-1390
(Rev. 5-93)

US DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE

ATTORNEY'S DOCKET NO.

H 2938 PCT/US

TRANSMITTAL LETTER TO THE UNITED STATES
DESIGNATED/ELECTED OFFICE (DO/EO/US)
CONCERNING A FILING UNDER 35 U.S.C. 371

U.S. APPLICATION NO. (if known sec. 17 CFR 1.5)

09/486677

INTERNATIONAL APPLICATION NO.

PCT/EP98/05355

INTERNATIONAL FILING DATE

August 24, 1998

PRIORITY DATE CLAIMED

September 1, 1997

TITLE OF INVENTION

COLD-STABLE FATTY ALCOHOL ALKOXYLATES

APPLICANT(S) FOR DO/EO/US

Ansgar Behler and Horst-Dieter Schares

Applicant herewith submits to the United States Designated/Elected Office (EO/DO/US) the following items and other information:

1. ☒ This is a **FIRST** submission of items concerning a filing under 35 U.S.C. 371.
 2. ☐ This a **SECOND** or **SUBSEQUENT** submission of items concerning a filing under 35 U.S.C. 371.
 3. ☐ This express request to begin national examination procedures (35 U.S.C. 371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39 (1).
 4. ☒ A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date.
 5. ☒ A copy of the International Application as filed (35 U.S.C. 371(c)(2)).
 - a. ☐ is transmitted herewith (required only if not transmitted by the International Bureau).
 - b. ☒ has been transmitted by the International Bureau.
 - c. ☐ is not required, as the application was filed in the United States Receiving Office (RO/US).
 6. ☒ A translation of the International Application into English (35 U.S.C. 371(c)(2)).
 7. ☒ Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3))
 - a. ☐ are transmitted herewith (required only if not transmitted by the International Bureau).
 - b. ☐ have been transmitted by the International Bureau.
 - c. ☐ have not been made; however, the time limit for making such amendments has NOT expired.
 - d. ☒ have not been made and will not be made.
 8. ☐ A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).
 9. ☒ An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)). **(UNEXECUTED)**
 10. ☐ A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).
- Items 11. to 16. below concern other document(s) or information included:**
11. ☐ An Information Disclosure Statement under 37 CFR 1.97 and 1.98.
 12. ☐ An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.
 13. ☐ A **FIRST** preliminary amendment
☐ A **SECOND** or **SUBSEQUENT** preliminary amendment.
 14. ☐ A substitute specification.
 15. ☐ A change of power of attorney and/or address letter.
 16. ☐ Other items or information.:

"Express Mail Post Office to Addressee" service Mailing Label Number**EL541611884US.**

U.S. Application No. (If known see 37 CFR 1.301) 09/486677	INTERNATIONAL APPLICATION NO. PCT/EP98/05355	ATTORNEY'S DOCKET NUMBER H 2938 PCT/US						
17. ■ The following fees are submitted: Basic National Fee (37 CFR 1.492(a)(1)-(6)): Search Report has been prepared by the EPO or JPO..... \$840.00 International preliminary examination fee paid to USPTO (37CFR 1.482) \$670.00 No international preliminary examination fee paid to USPTO (37 CFR 1.482) but international search fee paid to USPTO (37CFR 1.445(a)(2))..... \$690.00 Neither international preliminary examination fee (37CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO..... \$970.00 International preliminary examination fee paid to USPTO (37CFR 1.482) and all claims satisfied provisions of PCT Article 33(2)-(4)..... \$96.00 ENTER APPROPRIATE BASIC FEE AMOUNT =		CALCULATIONS PTO USE ONLY <table style="width:100%; border-collapse: collapse;"> <tr> <td style="width:10%;"></td> <td style="width:10%; text-align: right;">\$</td> <td style="width:40%; text-align: right;">840</td> <td style="width:10%; text-align: right;">00</td> <td style="width:30%;"></td> </tr> </table>			\$	840	00	
	\$	840	00					
Surcharge of \$130.00 for furnishing the oath or declaration later than <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date 37 (CFR 1.492(e)).		<table style="width:100%; border-collapse: collapse;"> <tr> <td style="width:10%;"></td> <td style="width:10%; text-align: right;">\$</td> <td style="width:40%; text-align: right;">0</td> <td style="width:10%; text-align: right;">00</td> <td style="width:30%;"></td> </tr> </table>			\$	0	00	
	\$	0	00					
Claims	Number filed	Number Extra	Rate					
Total Claims	9 - 20 =	0	0 X \$18.00	\$	00			
Independent Claims	1 - 3 =	0	0 X \$78.00	\$	00			
Multiple dependent claims (s)(if applicable)		0	+ \$260.00	\$	00			
TOTAL OF ABOVE CALCULATIONS =				\$	84000			
Reduction by ½ for filing by small entity, if applicable. Verified Small Entity statement must also be filed. (Note 37 CFR 1.9, 1.27, 1.28).				\$	000			
SUBTOTAL =				\$	84000			
Processing fee of \$130.00 for furnishing the English translation later the <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date (37CFR 1.492(f)).				\$	000			
TOTAL NATIONAL FEE =				\$	84000			
Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). \$40.00 per property				\$	000			
TOTAL FEES ENCLOSED =				\$	84000			
				Amount to be: refunded	\$-----			
				charged	\$840.00			

a. ☐ A check in the amount of \$_____ to cover the above fees is enclosed.

b. ■ Please charge my Deposit Account No. 50-1177 in the amount of \$840.00 to cover the above fees.
 A triplicate copy of this sheet is enclosed. Order No. 00-0090.

c. ■ The Assistant Commissioner is hereby authorized to charge any additional fees which may be required, or credit any
 overpayment to Deposit Account No. 50-1177. A triplicate copy of this sheet is enclosed.

**NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137 (a) or (b)) must be
 filed and granted to restore the application to pending status.**

SEND ALL CORRESPONDENCE TO: **Cognis Corporation, Law Dept.**
2500 Renaissance Blvd, Suite 200
Gulph Mills, PA 19406

SIGNATURE

Aaron R. Ettelman
NAME ATTORNEY FOR APPLICANT
42.516
REGISTRATION NUMBER

"Express Mail" mailing label number EL541612377US.

PATENT
Docket No. H 2938 PCT/US

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

RE: Serial No. 09/486,677
PCT/EP98/05355
International Filing Date: August 24, 1998
Priority Date Claimed: September 1, 1997
Applicant: Behler et al.
Title: COLD-STABLE FATTY ALCOHOL ALKOXYLATES
Applicants' Reference: H 2938 PCT/US

PRELIMINARY AMENDMENT

Commissioner for Patents
Box PCT
Washington, DC 20231

ATTN: DO/EO/US

Prior to the examination of the above-identified national stage application pursuant to the first submission under 35 U.S.C. §371 filed on March 1, 2000, please amend the English translation of the application submitted therewith, without prejudice, as follows:

In the Specification:

Please amend the instant Specification, without prejudice, as follows:

At page 1, please delete all text above line 7, and insert therefor the following:

--TITLE OF THE INVENTION

Random Fatty Alcohol Alkoxylates With Low Temperature Stability,
and Methods of Producing and Using The Same

BACKGROUND OF THE INVENTION--

At page 2, between lines 28 and 29 thereof, insert:

--BRIEF SUMMARY OF THE INVENTION--.

At page 3, between lines 1 and 2, insert the following text:

--The present invention includes low-temperature stable random polymers of fatty alcohols with ethylene oxide and propylene oxide, the ethylene oxide and propylene oxide being present in a certain selected quantity ratio. The present invention also includes a process for the production of such random polymers of fatty alcohols, and to methods of using such polymers as surfactants in water-dilutable concentrates including, for example, concentrates of detergents, pesticides and/or agrochemicals.--

At page 3, between lines 19 and 20 thereof, insert:

--DETAILED DESCRIPTION OF THE INVENTION--.

At page 6, line 14 thereof, replace "2EO" with --2PO--.

At page 7, line 17 thereof, replace "(3.72 moles)" with --(3.77 moles)--.

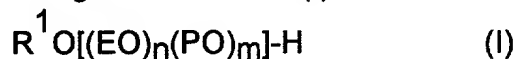
At page 8, between lines 1 and 2, insert --What is claimed is:--.

Please add new page 10, which is attached hereto, containing an Abstract of the Disclosure, following the claims.

In the Claims:

Please add new claims 10-30, as follow:

--10. (New) A mixture of polymers comprising random fatty alcohol alkoxylates according to the general formula (I):



wherein R^1 represents an alkyl group having from about 6 to about 22 carbon atoms, each EO represents $-CH_2CH_2O-$, each PO independently represents $-C(CH_3)HCH_2O-$ or $-CH_2C(CH_3)HO-$, and wherein n represents the average number of EO units present in each random fatty alcohol alkoxylate and has a value of from

about 2 to about 7, and wherein m represents the average number of PO units present in each random fatty alcohol alkoxylate and has a value of from about 1.5 to about 3.--

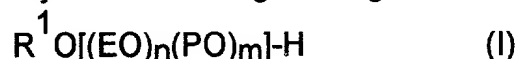
--11. (New) The mixture according to claim 10, wherein n has a value of from about 3 to about 5.--

--12. (New) The mixture according to claim 10, wherein m has a value of from about 2 to about 2.5.--

--13. (New) The mixture according to claim 10, wherein n has a value of from about 3 to about 5, and m has a value of from about 2 to about 2.5.--

--14. (New) The mixture according to claim 10, wherein R^1O represents a fatty alcohol residue derived from a fatty alcohol mixture, said mixture comprising at least about 30% by weight of alcohols having from about 14 to about 18 carbon atoms and up to about 70% by weight of alcohols having from about 6 to about 12 carbon atoms.--

--15. (New) A process for producing a mixture of polymers comprising random fatty alcohol alkoxylates according to the general formula (I) :



wherein R^1 represents an alkyl group having from about 6 to about 22 carbon atoms, each EO represents $-CH_2CH_2O-$, each PO independently represents $-C(CH_3)HCH_2O-$ or $-CH_2C(CH_3)HO-$, and wherein n represents the average number of EO units present in each random fatty alcohol alkoxylate and has a value of from about 2 to about 7, and wherein m represents the average number of PO units present in each random fatty alcohol alkoxylate and has a value of from about 1.5 to about 3; said process comprising reacting ethylene oxide, propylene oxide and fatty alcohol in the presence of an aqueous base, wherein the propylene oxide and the ethylene oxide are present during the reaction in a molar ratio of from about 10:90 to about 60:40.--

--16. (New) The process according to claim 15, wherein the

propylene oxide and the ethylene oxide are present during the reaction in a molar ratio of from about 10:90 to about 50:50.--

--17. (New) The process according to claim 15, wherein the propylene oxide and the ethylene oxide are present during the reaction in a molar ratio of from about 25:75 to about 50:50.--

--18. (New) The process according to claim 15, wherein the propylene oxide and the ethylene oxide are present during the reaction in a molar ratio of from about 25:75 to about 40:60.--

--19. (New) The process according to claim 15, wherein said fatty alcohol is a mixture of at least two fatty alcohols, said mixture comprising at least about 30% by weight of alcohols having from about 14 to about 18 carbon atoms and up to about 70% by weight of alcohols having from about 6 to about 12 carbon atoms.--

--20. (New) The process according to claim 15, wherein said aqueous base comprises a hydroxide selected from the group consisting of alkali metal hydroxides and alkali earth metal hydroxides.--

--21. (New) The process according to claim 20, wherein said hydroxide comprises potassium hydroxide.--

--22. (New) The product of the process according to claim 15.--

--23. (New) A water-dilutable concentrate comprising an active ingredient and a surfactant, said surfactant comprising a mixture of polymers comprising random fatty alcohol alkoxylates according to the general formula (I):



wherein R^1 represents an alkyl group having from about 6 to about 22 carbon atoms, each EO represents $-CH_2CH_2O-$, each PO independently represents $-C(CH_3)HCH_2O-$ or $-CH_2C(CH_3)HO-$, wherein n represents the average number of EO units present in each random fatty alcohol alkoxylate and has a value of from about 2 to about 7, and wherein m represents the average number of PO units

Case H 2938 PCT/US
Serial No. 09/486,677

present in each random fatty alcohol alkoxylate and has a value of from about 1.5 to about 3.--

--24. (New) The water-dilutable concentrate according to claim 23, wherein said active ingredient comprises one or more components selected from the group consisting of detergents, agrochemicals and pesticides.--

--25. (New) The water-dilutable concentrate according to claim 23, wherein said active ingredient comprises a detergent, and wherein said surfactant is present in an amount of from about 10% to about 30% by weight, based on the active ingredient.--

--26. (New) The water-dilutable concentrate according to claim 23, wherein said active ingredient comprises an agrochemical and/or a pesticide, and wherein said surfactant is present in an amount of from about 0.1% to about 15% by weight, based on the active ingredient.--

--27. (New) The water-dilutable concentrate according to claim 23, wherein n has a value of from about 3 to about 5.--

--28. (New) The water-dilutable concentrate according to claim 23, wherein m has a value of from about 2 to about 2.5.--

--29. (New) The water-dilutable concentrate according to claim 23, wherein n has a value of from about 3 to about 5, and m has a value of from about 2 to about 2.5.--

--30. (New) The water-dilutable concentrate according to claim 23, wherein R^1O represents a fatty alcohol residue derived from a fatty alcohol mixture, said mixture comprising at least about 30% by weight of alcohols having from about 14 to about 18 carbon atoms and up to about 70% by weight of alcohols having from about 6 to about 12 carbon atoms.--

Please cancel claims 1-9, without prejudice.

REMARKS

Claims 10-30 are currently pending in the instant application.

The Specification has been amended to include the preferred section headings pursuant to 37 C.F.R. §1.77. The Specification has also been amended to correct two obvious typographical errors at pages 6 and 7. An Abstract of the Disclosure in accordance with the abstract of the corresponding international publication has been added on a separate sheet following the claims. It is submitted that the amendments to the Specification made herein introduce no new matter. Their entry is therefore proper and respectfully requested.

Original claims 1-9 have been canceled and replaced with new claims 10-30 in an effort to place the claims in more proper U.S. format, with increased clarity, for the purposes of particularly pointing out and distinctly claiming that which Applicants regard as their invention. New claims 10-30 are supported by the claims as originally filed and in the Specification, for example, at page 3, lines 2-19, at page 4, lines 4-8, and 24-29, at page 5, lines 13-15, and at page 5, line 24 through page 6, line 11. No new matter has been introduced. Entry is therefore proper and respectfully requested.

Case H 2938 PCT/US
Serial No. 09/486,677

Prompt examination of the instant application in view of the amendments made
herein is respectfully requested.

Respectfully submitted,

ANSGAR BEHLER, et al.

June 26, 2000
(Date)

A. R. Ettelman
AARON R. ETTELMAN
(Reg. No. 42,516)
Attorney for Applicants
Telephone: (610) 278-4930
Facsimile: (610) 278-6548
E-Mail: AARON.ETTELMAN@HENKEL-AMERICAS.COM

Cognis Corporation, Patent Dept.
2500 Renaissance Blvd., Suite 200
Gulph Mills, PA 19406

ARE/ras

G:\DATA\AMEND\H2938.PAM

"Express Mail" mailing label number EL541612377US .

PATENT
Docket No. H 2938 PCT/US

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

RE: Serial No. 09/486,677
PCT/EP98/05355
International Filing Date: August 24, 1998
Priority Date Claimed: September 1, 1997
Applicant: Behler et al.
Title: COLD-STABLE FATTY ALCOHOL ALKOXYLATES
Applicants' Reference: H 2938 PCT/US

PRELIMINARY AMENDMENT

Commissioner for Patents
Box PCT
Washington, DC 20231

ATTN: DO/EO/US

Prior to the examination of the above-identified national stage application pursuant to the first submission under 35 U.S.C. §371 filed on March 1, 2000, please amend the English translation of the application submitted therewith, without prejudice, as follows:

In the Specification:

Please amend the instant Specification, without prejudice, as follows:

At page 1, please delete all text above line 7, and insert therefor the following:

--TITLE OF THE INVENTION

Random Fatty Alcohol Alkoxylates With Low Temperature Stability,
and Methods of Producing and Using The Same

BACKGROUND OF THE INVENTION--

At page 2, between lines 28 and 29 thereof, insert:

--BRIEF SUMMARY OF THE INVENTION--.

Cold-stable Fatty Alcohol Alkoxylates

This invention relates to low-temperature-stable random polymers of fatty alcohols with ethylene oxide and propylene oxide, the ethylene oxide and propylene oxide being present in a certain selected quantity ratio, to a process for the production of such compounds and to the use of such compounds as surfactants in water-dilutable concentrates, more particularly in concentrates of detergents, pesticides and agrochemicals.

Alcohol alkoxylates, which are also known as alkyl polyglycol ethers, are compounds which have been known for decades and which are obtained by reacting alcohols with ethylene oxide and/or propylene oxide.

The reaction takes place at elevated temperatures and pressures in the presence of acidic or alkaline catalysts. The use of basic compounds of the alkali and alkaline earth metals is of particular practical significance for the alkoxylation of fatty alcohols. Suitable basic compounds are alkali metal and alkaline earth metal alkoxylates, such as sodium methylate and potassium methylate, or alkali metal and alkaline earth metal hydroxides, such as sodium hydroxide and potassium hydroxide. Recently, alkali metal and alkaline earth metal hydroxides have preferably been used as basic catalysts because, where alkali metal and alkaline earth metal alkoxylates are used, organic solvents, such as methanol, are essential. By contrast, where the alkali metal and alkaline earth metal hydroxides are used, the reaction gives good yields, even in aqueous medium.

However, where aqueous solutions of alkali metal and alkaline earth metal hydroxides are used as basic catalysts, products with unfavorable low-temperature behavior which often tend to precipitate, even at room temperature, are obtained, particularly in the ethoxylation of the fatty alcohols. This is attributable inter alia to the fact that the presence of

aqueous solutions of alkali metal and alkaline earth metal hydroxides promotes the formation of high molecular weight polyethylene glycols as secondary products. Although, in principle, these polyethylene glycols can be removed by extraction with suitable solvents, such as water, this does
5 involve another process step which is very time-consuming and, in addition, cannot be universally applied.

According to the abstract of **Japanese patent application 07,303,825** from the **Journal CA Selects in: Alkoxylated Oleochemicals, Issue 1996, page 5**, published by the American Chemical Society,
10 Columbus, Ohio, No. 124, random adducts of C₈₋₁₈ alcohols containing 5 to 15 moles of ethylene oxide and 0.3 to 5.0 moles of propylene oxide show improved flow behavior at low temperatures. In the only example cited, lauryl alcohol is reacted with about 9 moles of ethylene oxide and about 2.4 moles of propylene oxide in the presence of potassium hydroxide as basic
15 catalyst, a product with a pour point of 7.5°C being obtained. However, this product also tends to precipitate in storage at temperatures below 0°C which is problematical.

The problem addressed by the present invention was to provide fatty alcohol alkoxylates which would show very favorable low-temperature
20 behavior and would not have any tendency to precipitate. The low-temperature behavior would be so pronounced that the products would remain clear liquids even at temperatures below 0°C so that no precipitation would occur, even in the event of storage at low temperatures. In addition, the products obtained would be readily soluble in cold water so
25 that they would be suitable for use as surface-active compounds, for example in liquid concentrates of detergents. In addition, the compounds would lend themselves to production in the presence of aqueous solutions of bases.

The problem stated above has surprisingly been solved by random
30 polymers of fatty alcohols which contain ethylene oxide and propylene

oxide in random-polymerized form in a certain selected ratio.

Accordingly, the present invention relates to random polymers of fatty alcohols with ethylene oxide and propylene oxide corresponding to formula (I):

5



10 in which R^1 is an alkyl group containing 6 to 22 carbon atoms, EO stands $\text{CH}_2\text{CH}_2\text{O}$, PO stands for $\text{CHCH}_3\text{CH}_2\text{O}$ and/or $\text{CH}_2\text{CHCH}_3\text{O}$, n is a whole or broken number of 2 to 7 and m is a whole or broken number of 1.5 to 3, characterized in that the molar ratio of propylene oxide to ethylene oxide is from 10:90 to 50:50.

15 Random polymers of fatty alcohols corresponding to formula (I), in which the molar ratio of propylene oxide to ethylene oxide is from 25:75 to 40:60, are particularly preferred.

In addition, random polymers of fatty alcohols corresponding to formula (I), in which n is a whole or broken number of 3 to 5, and random polymers of formula (I), in which m is a whole or broken number of 2 to 2.5, are preferred.

20 R^1 in formula (I) is derived from fatty alcohols with the formula R^1OH which contain 6 to 22 carbon atoms. Fatty alcohols in this context are understood to be primary aliphatic alcohols in which R^1 is an aliphatic, linear or branched hydrocarbon radical containing 6 to 22 carbon atoms and 0 and/or 1, 2 or 3 double bonds. Typical examples are caproic alcohol, 25 caprylic alcohol, 2-ethylhexyl alcohol, capric alcohol, lauryl alcohol, isotridecyl alcohol, myristyl alcohol, cetyl alcohol, palmitoleyl alcohol, stearyl alcohol, isostearyl alcohol, oleyl alcohol, elaidyl alcohol, petroselinyl alcohol, linolyl alcohol, linolenyl alcohol, elaeostearyl alcohol, arachyl alcohol, gadoleyl alcohol, behenyl alcohol, erucyl alcohol and brassidyl 30 alcohol and the technical mixtures thereof obtained, for example, in the

high-pressure hydrogenation of technical methyl esters based on fats and oils or aldehydes from Roelen's oxosynthesis and as monomer fraction in the dimerization of unsaturated fatty alcohols.

According to the invention, technical fatty alcohols containing at
5 least 30% by weight C_{14-18} fatty alcohols and at most 70% by weight C_{6-12} fatty alcohols are preferred. The percentages by weight are based on the fatty alcohol mixture. Examples of such fatty alcohol mixtures are coconut, palm, palm kernel or tallow fatty alcohol.

The random polymers according to the invention are distinguished
10 by very good low-temperature behavior, i.e. they are free-flowing and preferably clear even at temperatures below 0°C . The low-temperature behavior is determined by measurement of the cold cloud point in accordance with DIN ISO 3015. The products according to the invention are also distinguished by very good solubility in cold water, i.e. they have
15 very short dissolving times. Dissolving behavior was determined by measuring the time which 10 g of the compound takes to form an optically clear solution in 90 g of stirred deionized water (temperature 23°C).

The present invention also relates to a process for the production of random polymers of fatty alcohols corresponding to formula (I) by reacting
20 ethylene oxide and propylene oxide with fatty alcohols corresponding to the formula $R^1\text{OH}$ in the presence of aqueous bases, characterized in that propylene oxide and ethylene oxide in a molar ratio of 10:90 to 40:60 are reacted with fatty alcohols in known manner.

The random polymerization is a process known per se in which the
25 above-described alcohols $R^1\text{OH}$ are reacted with propylene and ethylene oxide in a pressure vessel at temperatures of 120 to 190°C under a pressure of 3 to 5 bar. According to the invention, aqueous solutions of alkali metal and/or alkaline earth metal hydroxides, for example potassium hydroxide, are used as the basic compounds. The hydroxides are normally
30 used in quantities of 0.2 to 5% by weight and preferably in quantities of 0.3

to 1.5% by weight, expressed as hydroxide and based on the mixture as a whole. It is advisable to use the hydroxides in the form of 40 to 60% by weight aqueous solutions. Propylene and ethylene oxide are reacted together with the fatty alcohols. To this end, they may be separately introduced into the reaction vessel through two different nozzles or may even be premixed in a mixing vessel and then introduced into the reactor. It is crucial to the invention that propylene oxide and ethylene oxide are used in the quantities and mixing ratios mentioned above. In general, ethylene oxide is used in a quantity of 2 to 7 moles and preferably 3 to 5 moles per mole fatty alcohol while propylene oxide is used in a quantity of 1.5 to 3 moles and preferably 2 to 2.5 moles per mole of fatty alcohol. A molar mixing ratio of propylene oxide to ethylene oxide of preferably 25:75 to 40:60 is of particular significance. The reaction of ethylene oxide and propylene oxide together with the fatty alcohols gives so-called random polymers, i.e. the addition takes place in statistical distribution.

If desired, the process according to the invention may be followed by neutralization of the basic catalyst. The neutralization step may be carried out with inorganic and/or organic acids, such as lactic acid, oxalic acid, citric acid, acetic acid, phosphoric acid or methane sulfonic acid. Neutralization to values of 6.5 to 7.5 is generally advisable when unsaturated fatty alcohols are used because they easily discolor under the influence of atmospheric oxygen. The products are obtained in the form of a 100% clear liquid.

The present invention also relates to the use of the above-described random polymers of fatty alcohols as surface-active agents in water-dilutable concentrates, for example in concentrates of detergents or in concentrates of pesticides and agrochemicals. In recent years, detergents, for example, have been increasingly marketed as concentrates which contain only a little water. Such concentrates can be diluted with water before use by the end user. The concentrates are expected to dissolve

quickly without forming any precipitates. This is achieved by using the compounds according to the invention as surface-active agent. The products according to the invention may be used in detergent concentrates in quantities of 10 to 30% by weight, based on active ingredient in the concentrate. The concentrates may additionally contain typical constituents, such as anionic surfactants, nonionic surfactants, preservatives, solubilizers, etc., which are typical of detergents, in the usual quantities.

The products according to the invention may be used in pesticide and agrochemical concentrates in quantities of 0.1 to 15% by weight, based on active principle in the concentrate. In this case, too, the concentrates may contain typical constituents in typical quantities.

Examples

1. Production of a random polymer of a $C_{12/14}$ alcohol+5EO+2EO

366.3 g (1.89 moles) of a C_{12}/C_{14} fatty alcohol mixture (about 40% C_{12} and 60% by weight C_{14}) were introduced into a pressure vessel with 5 g of a 50% by weight aqueous potassium hydroxide solution. The vessel was then evacuated for 30 minutes at 100°C and subsequently purged with nitrogen. A mixture of 414.9 g (9.43 moles) of ethylene oxide and 218.8 g (3.77 moles) of propylene oxide was then introduced at 120°C. The maximum pressure was 5 bar. On completion of the reaction, the mixture was stirred for 1 hour at 120°C and the reactor evacuated for another 30 minutes at 120°C. The product obtained was neutralized with lactic acid.

A clear liquid product with a cold cloud point of -4°C was obtained; the dissolving time in water was 5 seconds.

2. Production of a random polymer of a $C_{12/14}$ alcohol+3EO+2PO

As in Example 1, 439.1 g (2.26 moles) of a C_{12}/C_{14} fatty alcohol were reacted with a mixture of 298.5 g (6.78 moles) of ethylene oxide and 262.4 g (4.52 moles) of propylene oxide in the presence of 5 g of a 50% by

weight aqueous potassium hydroxide solution.

A clear liquid product with a cold cloud point of -14°C which took 3 seconds to dissolve in water was obtained.

5 Comparison Example 1: Block polymer of a $\text{C}_{12/14}$ alcohol with 1PO+5EO+1PO

 In accordance with Example 1, the quantities of fatty alcohol described in Example 1 were reacted first at 120°C with 109.4 g (1.89 moles) of propylene oxide, then at 180°C with 414.9 g (9.43 moles) of ethylene oxide and, on completion of the reaction, at 120°C with another
10 109.4 g (1.89 moles) of propylene oxide.

 A clear liquid product with a cold cloud point of 1.5°C was obtained. The dissolving time in water was 75 seconds.

 Comparison Example 2: Block polymer of a $\text{C}_{12/14}$ alcohol with 5EO+2PO

15 In accordance with Example 1, the quantity of fatty alcohol described in Example 1 was reacted first at 180°C with 414.9 g (9.43 moles) of ethylene oxide and then at 120°C with 218.8 g (3.72 moles) of propylene oxide.

 A clear liquid product with a cold cloud point of 7.5°C was obtained.
20 The dissolving time in water was 8 seconds.

CLAIMS

1. Random polymers of fatty alcohols with ethylene oxide and propylene oxide corresponding to formula (I):



in which R^1 is an alkyl group containing 6 to 22 carbon atoms, EO stands CH_2CH_2O , PO stands for $CHCH_3CH_2O$ and/or CH_2CHCH_3O , n is a whole or broken number of 2 to 7 and m is a whole or broken number of 1.5 to 3,
10 characterized in that the molar ratio of propylene oxide to ethylene oxide is from 10:90 to 50:50.

2. Random polymers as claimed in claim 1, characterized in that the molar ratio of propylene oxide to ethylene oxide is from 25:75 to 40:60.

3. Random polymers as claimed in claim 1, characterized in that n is a
15 whole or broken number of 3 to 5.

4. Random polymers as claimed in claim 1, characterized in that m is a whole or broken number of 2 to 2.5.

5. Random polymers as claimed in claim 1, characterized in that R^1 is derived from a fatty alcohol mixture containing at least 30% by weight of
20 C_{14-18} fatty alcohols and at most 70% by weight of C_{6-12} fatty alcohols.

6. A process for the production of random polymers of fatty alcohols with ethylene oxide and propylene oxide corresponding to formula (I):

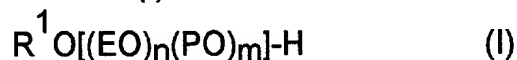


with the definitions given in claim 1, by reacting ethylene oxide and propylene oxide with fatty alcohol having the formula R^1OH in the presence of aqueous bases, characterized in that the propylene oxide and ethylene oxide in a molar ratio of 10:90 to 40:60 are reacted with fatty alcohols by
30 methods known per se.

7. A process as claimed in claim 6, characterized in that the molar ratio of propylene oxide to ethylene oxide is in the range from 25:75 to 40:60.
8. A process as claimed in claim 6, characterized in that a fatty alcohol mixture containing at least 30% by weight of C₁₄₋₁₈ fatty alcohols and at
5 most 70% by weight of C₆₋₁₂ fatty alcohols is reacted.
9. The use of the random polymers of fatty alcohols corresponding to formula (I) claimed in claim 1 as a surfactant in water-dilutable concentrates.

ABSTRACT OF THE DISCLOSURE

Random fatty alcohol alkoxylates with low temperature stability which are polymerization products of fatty alcohols, ethylene oxide and propylene oxide, according to the general formula (I) :



wherein R^1 represents an alkyl group having from about 6 to about 22 carbon atoms, each EO represents $-CH_2CH_2O-$, each PO independently represents $-C(CH_3)HCH_2O-$ or $-CH_2C(CH_3)HO-$, wherein n represents the average number of EO units present in each random fatty alcohol alkoxylate and has a value of from about 2 to about 7, and wherein m represents the average number of PO units present in each random fatty alcohol alkoxylate and has a value of from about 1.5 to about 3; are disclosed. A process for the production of such random fatty alcohol alkoxylates and methods of their use in water-dilutable concentrates are also disclosed.

Type a plus sign (+) inside this box → ☐

0010/PTO Rev. 6/95 DECLARATION FOR UTILITY OR DESIGN PATENT APPLICATION <input type="checkbox"/> Declaration Submitted with Initial Filing OR <input checked="" type="checkbox"/> Declaration Submitted after Initial Filing	U.S. Department of Commerce Patent and Trademark Office	Attorney Docket Number	H 2938 PCT/US
		First Named Inventor	BEHLER, Ansgar
	COMPLETE IF KNOWN		
	Application Number	09/486,677	
	Filing Date	06/26/2000	
	Group Art Unit		
		Examiner Name	

As a below named inventor, I hereby declare that:

My residence, post office address, and citizenship are as stated below next to my name.

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled:

COLD-STABLE FATTY ALCOHOL ALKOXYLATES

(Title of the Invention)

the specification of which

☐ is attached hereto

OR

☒ was filed on (MM/DD/YYYY) 08/24/1998 as United States Application Number or PCT International

Application Number PCT/EP98/05355 and was amended on (MM/DD/YYYY) (if applicable).

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment specifically referred to above.

I acknowledge the duty to disclose information which is material to patentability as defined in Title 37 Code of Federal Regulations, § 1.56.

I hereby claim foreign priority benefits under Title 35, United States Code §119(a)-(d) or §365(b) of any foreign application(s) for patent or inventor's certificate, or §365(a) of any PCT International application which designated at least one country other than the United States of America, listed below and have also identified below, by checking the box, any foreign application for patent or inventor's certificate, or of any PCT International application having a filing date before that of the application on which priority is claimed.

Prior Foreign Application Number(s)	Country	Foreign Filing Date (MM/DD/YYYY)	Priority Not Claimed	Certified Copy Attached? YES NO	
197 38 108.1	Germany	09/01/1997	<input type="checkbox"/>	<input type="checkbox"/>	<input checked="" type="checkbox"/>
			<input type="checkbox"/>	<input type="checkbox"/>	<input type="checkbox"/>
			<input type="checkbox"/>	<input type="checkbox"/>	<input type="checkbox"/>
			<input type="checkbox"/>	<input type="checkbox"/>	<input type="checkbox"/>
			<input type="checkbox"/>	<input type="checkbox"/>	<input type="checkbox"/>
			<input type="checkbox"/>	<input type="checkbox"/>	<input type="checkbox"/>
			<input type="checkbox"/>	<input type="checkbox"/>	<input type="checkbox"/>

☐ Additional foreign application numbers are listed on a supplemental priority sheet attached hereto:

I hereby claim the benefit under Title 35, United States Code §119(e) of any United States provisional application(s) listed below.

Application Number(s)	Filing Date (MM/DD/YYYY)	Additional provisional application numbers are listed on a supplemental priority sheet attached hereto.
		<input type="checkbox"/>

Burden Hour Statement: This form is estimated to take .4 hours to complete. Time will vary depending upon the needs of the individual case. Any comments on the amount of time you are required to complete this form should be sent to the Chief Information Officer, Patent and Trademark Office, Washington, DC 20231. DO NOT SEND FEES OR COMPLETED FORMS TO THIS ADDRESS. SEND TO: Assistant Commissioner for Patents, Washington, DC 20231.

**"Express Mail Post Office
to Addressee" service
Mailing Label Number
EL541612377US**

DECLARATION

Page 2

I hereby claim the benefit under Title 35, United States Code §120 of any United States application(s), or §365(c) of any PCT international application designating the United States of America, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States or PCT international application in the manner provided by the first paragraph of Title 35, United States Code §112.1 acknowledge the duty to disclose information which is material to patentability as defined in Title 37, Code of Federal Regulations §1.56 which became available between the filing date of the prior application and the national or PCT international filing date of this application.

U.S. Parent Application Number	PCT Parent Number	Parent Filing Date (MM/DD/YYYY)	Parent Patent Number (if applicable)
	PCT/EP98/05355	08/24/1998	

☐ Additional U.S. or PCT international application numbers are listed on a supplemental priority sheet attached hereto.

As a named inventor, I hereby appoint the following attorney(s) and/or agent(s) to prosecute this application and to transact all business in the Patent and Trademark Office connected therewith:

☐ Firm Name Customer Number or label

☒ List Attorney(s) and/or agent(s) name and registration number below:

Name	Registration Number	Name	Registration Number
John E. Drach	32,891	Henry E. Millson, Jr.	18,980
Steven J. Trzaska	36,296		
Aaron R. Ettelman	42,516		

☐ Additional attorney(s) and/or agent(s) named on a supplemental sheet attached hereto.

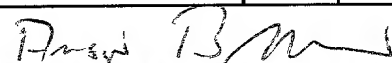
Please direct all correspondence to: ☐ Customer Number or label OR ☒ Fill in correspondence address below

Name	Aaron R. Ettelman				
Address	Cognis Corporation - Patent Department				
Address	2500 Renaissance Boulevard, Suite 200				
City	Gulph Mills	State	PA	ZIP	19406
Country	USA	Telephone	610-278-4930	Fax	610-278-6548

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Name of Sole or First Inventor: ☐ A petition has been filed for this

Given Name	Ansgar	Middle Initial		Family Name	Behler	Suffix e.g. Jr.	
------------	--------	----------------	--	-------------	--------	-----------------	--

Inventor's Signature		Date	03/23/2000
----------------------	---	------	------------

Residence: City	Bottrop	State		Country	Germany	Citizenship	Germany
-----------------	---------	-------	--	---------	---------	-------------	---------

Post Office Address	Siegfriedstrasse 80				
---------------------	---------------------	--	--	--	--

Post Office Address					
---------------------	--	--	--	--	--

City	46240 Bottrop	State		Zip		Country	Germany	Applicant Authority	
------	---------------	-------	--	-----	--	---------	---------	---------------------	--

☒ Additional inventors are being named on supplemental sheet(s) attached hereto

DECLARATION**ADDITIONAL INVENTOR(S)
Supplemental Sheet**

Name of Additional Joint Inventor, if any:

☐

A petition has been filed for this unsigned inventor

Given Name	Horst-Dieter	Middle Initial		Family Name	Schaes	Suffix e.g. Jr.	
Inventor's Signature	<i>Horst-Dieter</i>					Date	<i>03/23/2000</i>
Residence: City	Erkrath	State	DEX	Country	Germany	Citizenship	Germany
Post Office Address	Naheweg 11						
Post Office Address							
City	40699 Erkrath	State		Zip		Country	Germany
						Applicant Authority	

Name of Additional Joint Inventor, if any:

☐

A petition has been filed for this unsigned inventor

Given Name		Middle Initial		Family Name		Suffix e.g. Jr.	
Inventor's Signature						Date	
Residence: City		State		Country		Citizenship	
Post Office Address							
Post Office Address							
City		State		Zip		Country	
						Applicant Authority	

Name of Additional Joint Inventor, if any:

☐

A petition has been filed for this unsigned inventor

Given Name		Middle Initial		Family Name		Suffix e.g. Jr.	
Inventor's Signature						Date	
Residence: City		State		Country		Citizenship	
Post Office Address							
Post Office Address							
City		State		Zip		Country	
						Applicant Authority	

Name of Additional Joint Inventor, if any:

☐

A petition has been filed for this unsigned inventor

Given Name		Middle Initial		Family Name		Suffix e.g. Jr.	
Inventor's Signature						Date	
Residence: City		State		Country		Citizenship	
Post Office Address							
Post Office Address							
City		State		Zip		Country	
						Applicant Authority	

☐

Additional inventors are being named on supplemental sheet(s) attached hereto